

1300 Eye Street N.W.
Washington, DC 20005
Telephone: 202 659.0100
Facsimile: 202.659.0105
www.novakdruce.com

**Novak Druce DeLuca
& Quigg, LLP**

Fax

**RECEIVED
CENTRAL FAX CENTER**

APR 05 2006

Certificate

To: USPTO - Mail Stop
Certificate of Corrections Branch **From:** Jason D. Voight

Fax: (571) 273-8300 **Pages:** 5

Phone: (571) 272-1000 **Date:** April 5, 2006

Re: Attorney Docket: 50721 **cc:**

APR 10 2006

of Correction

☐ Urgent ☐ For Review ☐ Please Comment ☐ Please Reply ☐ Please Recycle

• **Comments:**

In re Application of: **MAAS et al.**

Serial No.: **09/937,815**

Patent No.: **6,844,290**

Issue Date: **January 18, 2005**

Title: **OLIGOMERISATION CATALYST**

Attachments: **Request for Certificate of Correction
Certificate of Correction Form PTO/SB/44 (PTO-1050)
Mark-up copy of Letters Patent**

CONFIDENTIALITY NOTICE: This facsimile and any attachments contain information from the law firm of Novak Druce DeLuca & Quigg, LLP, which may be confidential and/or privileged. The information is intended to be for the use of the individual or entity named on this facsimile. If you are not the intended recipient, be aware that any disclosure, copying, distribution or use of the contents of this facsimile is prohibited. If you receive this facsimile in error, please notify us by reply facsimile immediately so that we can arrange for the retrieval of the original documents at no cost to you.

APR 10 2006

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: MAAS et al.

Art Unit: 1713

Patent No.: 6,844,290 *B1*

Examiner: C. Caixia LU

Issued: January 18, 2005

Confirmation No.: 5442

For: OLIGOMERISATION CATALYST

Attorney Docket.: 50721

RECEIVED
CENTRAL FAX CENTER

APR 05 2006

Mail Stop Certificate of Correction
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

CERTIFICATE OF FACSIMILE

I hereby certify that this correspondence is being transmitted by
facsimile to U.S. Patent and Trademark Office fax no. (571) 273-
8300 on APR 05, 2006
Signature: *Michelle Bryant*

REQUEST FOR CERTIFICATE OF CORRECTION

Sir:

Applicants herewith submit a Certificate of Correction Form PTO/SB/44. It is respectfully requested that the Request for Certificate of Correction be entered.

The changes noted on the Certificate of Correction Form PTO/SB/44 correct the errors which occurred on the part of the U.S. Patent and Trademark Office. No fee should therefore be required.

It is not believed that a fee is required for filing of this paper. However, please charge any shortage in fees due in connection with the filing of this paper to Deposit Account No. 14.1437. Please credit any excess fees to such deposit account.

Respectfully submitted,

Jason D. Voight

Jason D. Voight
Registration No.: 42,205

Date: April 5, 2006
Novak Druce DeLuca & Quigg, LLP
1300 I Street, N.W., 400 East Tower
Washington, DC 20005
Telephone (202) 204-4746
Facsimile: (202) 659-0105

PTO/SB/44 (04-05)

Approved for use through 04/30/2007. OMB 0651-0033

U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number. (Also Form PTO-1050)

**UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION**

Page 1 of 1

PATENT NO. : 6,844,290 *B1*
APPLICATION NO. : 09/937,815
ISSUE DATE : January 18, 2005
INVENTOR(S) : MAAS et al.

It is certified that an error appears or errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

On the cover page at subsection (63) delete "Continuation-in-part of Application No. 09/277,823, filed on Mar. 29, 1999, now abandoned" and replace with the following paragraph

—The patent claims priority of U.S. application Serial No. 09/277,823, filed March 29, 1999, in accordance with PCT Article 8 and 35 U.S.C. § 119(a) and (b). —

Claim 2, column 15, indicated line 63

"C₇-C₈-arylalkyl" should read — C₇-C₈-arylalkyl —.

Claim 12, column 18, indicated line 1

"p position" should read —β position—.

MAILING ADDRESS OF SENDER (Please do not use customer number below):

Jason D. Voight
NOVAC DRUCE DELUCA & QUIGG LLP
1300 I Street, N.W., Suite 400 East Tower
Washington, DC 20005

This collection of information is required by 37 CFR 1.322, 1.323, and 1.324. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1.0 hour to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Attention Certificate of Corrections Branch, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

US006844290B1

(12) **United States Patent**
Maas et al. ✓(10) Patent No.: **US 6,844,290 B1**
(45) Date of Patent: **Jan. 18, 2005**(54) **OLIGOMERIZATION CATALYST** ✓(75) Inventors: **Helko Maas**, Schiffersstadt (DE) ✓
Shahram Mihan, Ludwigshafen (DE) ✓
Randolf Köhn, Bath (GB) ✓, **Guido Seifert**, Berlin (DE) ✓, **Jürgen Tropsch**, Römerberg (DE) ✓(73) Assignee: **BASF Aktiengesellschaft**, Ludwigshafen (DE) ✓

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days. ✓

(21) Appl. No.: **09/937,815** ✓(22) PCT Filed: **Mar. 25, 2000** ✓(86) PCT No.: **PCT/EP00/02660** ✓

§ 371 (c)(1).

(2), (4) Date: **Sep. 28, 2001** ✓(87) PCT Pub. No.: **WO00/58319** ✓PCT Pub. Date: **Oct. 5, 2000** ✓

Related U.S. Application Data

✓ (63) **The Patent Claims Priority of U.S. Appn. Serial No. 09/277,823**
Continuation-in-part of application No. 09/277,823, filed on Mar. 29, 1999, now abandoned.
U.S.C. 119(a) and (b). Filed March 29, 1999 in accordance with PCT Article 8 and 35
(30) Foreign Application Priority DataMay 14, 1999 (DE) 199 22 048
Sep. 11, 1999 (DE) 199 43 544(51) Int. Cl.⁷ **C07F 31/18; C07F 11/00**(52) U.S. Cl. **502/167; 502/103; 502/123; 526/159; 526/172**(58) Field of Search **502/103, 123, 502/167; 526/159, 172, 160, 165**(56) **References Cited****U.S. PATENT DOCUMENTS**

5,750,816 A 5/1998 Araki et al. 585/512

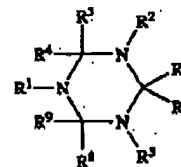
FOREIGN PATENT DOCUMENTS

EP 780 353 6/1997

Primary Examiner—Caixia Lu

(74) Attorney, Agent, or Firm—**Kell & Weinkauff**(57) **ABSTRACT**

An oligomerization catalyst for olefins is obtainable from a) a chromium compound CrX₃ and the at least equimolar amount, based on the chromium compound CrX₃, of a ligand L or from an existing chromium complex CrX₃L, in which the groups X are, independently of one another, abstractable counterions and L is a 1,3,5-triazacyclobexane of the formula I



where the groups R¹ to R⁶ are, independently of one another: hydrogen or organosilicon or substituted or unsubstituted carboorganic groups having from 1 to 30 carbon atoms, where two geminal or vicinal radicals R¹ to R⁶ may also be joined to form a five- or six-membered ring, and

b) at least one activating additive ✓

and also a process for preparing oligomers of olefins using these catalysts, the oligomers thus obtainable, and the oxo alcohols obtainable from these oligomers. ✓

13 Claims, No Drawings
✓ ✓

US 6,844,290 B1

15

16

TABLE 3

Data for Examples 23-26								
Ex.	Amount of chromium complex 10		Atom ratio	Atom ratio	T	Monomer	DMP ²⁾	
	[mg]	(μ mol)	Al:Cr	B ¹⁾ :Cr	[° C.]		[μ mol]	[min]
23	19.5	29.3	300:1	—	40	C ₂ H ₄ ⁴⁾	—	60
24	28.3	42.5	50:1 ⁵⁾	5.3:1	40	1-butene ⁴⁾	127.5	60
25	12.1	18.2	500:1	—	50	ethene	—	30
26	55	83	300:1	—	40	200 ml 1-butene	—	60

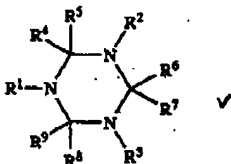
Ex.	Polymer [g]	Product [g]					Activity of the catalyst used [kg/(mol Cr·h)]				
		C ₆	C ₁₀	C ₁₄	C ₁₈	Polymer	C ₆	C ₁₀	C ₁₄	C ₁₈	Total
23	0.43	9.02	10.9	2.06	0.16	14.7	308	372	70	5.5	770
24	—	C ₁₀ : 2.31					—	C ₁₂ : 54.3			
25	0.07	6.63	1.96	—	—	3.8	730	215	0	0	945
26	—	C ₁₀ : 0.24, C ₁₂ : 2.09					—	C ₁₀ : 2.9, C ₁₂ : 25.2			

¹⁾Activation takes place by addition of DMAB and TIBAL.²⁾Triethylaluminum was used instead of TIBAL.³⁾2,5-Dimethylpyrrole.⁴⁾Gas was passed through.⁵⁾Use of triethylaluminum.

We claim:

1. A catalyst obtained from

- a) a chromium compound CrX₃ and the at least equimolar amount, based on the chromium compound CrX₃, of a ligand L or from an existing chromium complex CrX₃L, in which the groups X are, independently of one another, abstractable counterions and L is a 1,3,5-triazacyclohexane of the formula I



where the groups R¹ to R⁶ are, independently of one another, hydrogen or organosilicon or substituted or unsubstituted carbocyclic groups having from 1 to 30 carbon atoms, where two geminal or vicinal radicals R¹ to R⁶ may also be joined to form a five- or six-membered ring, and

- b) at least one activating additive selected from the group consisting of (i) and (ii) wherein:

- i) is a combination of an unsubstituted or substituted five-membered aromatic N-heterocycle and at least one aluminum alkyl, wherein some of the alkyl groups of the aluminum alkyl are optionally replaced by halogen and/or alkoxy, and

ii) is an alkylaluminum compound.

2. The catalyst defined in claim 1, wherein the groups R¹, R² and R³ in the 1,3,5-triazacyclohexane I are, independently of one another, substituted or unsubstituted C₁-C₁₂-alkyl, C₆-C₁₀-aryl or C₇-C₈-aryllalkyl.

3. The catalyst defined in claim 1, wherein the groups R¹, R² and R³ in the 1,3,5-triazacyclohexane I are, independently of one another, substituted or unsubstituted C₁-C₁₂-alkyl or C₇-C₈-aryllalkyl.

4. [(1,3,5-Tris(2-n-propylheptyl)-1,3,5-triazacyclohexane) CrCl₃].

5. [(1,3,5-Tris(2-ethylhexyl)-1,3,5-triazacyclohexane) CrCl₃].

6. A process for preparing oligomers having up to 30 carbon atoms by reaction of an olefin or a mixture of olefins at from 0 to 150° C. and pressures of from 1 to 200 bar in the presence of the catalyst defined in claim 1.

7. The catalyst defined in claim 1, wherein the groups R⁴, R⁵, R⁶, R⁷, R⁸ and R⁹ in the 1,3,5-triazacyclohexane I are, independently of one another, hydrogen or methyl.

8. A process as claimed in claim 6, wherein the olefin or mixture of olefins is selected from straight-chain and branched α -olefins having from 2 to 4 carbon atoms.

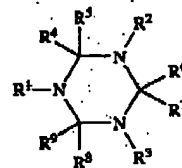
9. A process as claimed in claim 6, wherein the olefin or mixture of olefins is selected from 1-butene and 1-butene in mixture with its isomers.

10. A process as claimed in claim 6, wherein the olefin or mixture of olefins is employed in form of a raffinate comprising 1-butene in mixture with its isomers.

11. A process as claimed in claim 6, wherein the olefin is ethene.

12. A process as claimed in claim 6, wherein the catalyst is obtained from

- a) a chromium compound CrX₃ and the at least equimolar amount, based on the chromium compound CrX₃, of a ligand L or from an existing chromium complex CrX₃L, in which the groups X are, independently of one another, abstractable counterions and L is a 1,3,5-triazacyclohexane of the formula I



US 6,844,290 B1

17

where the groups R^1 to R^3 are, independently of one another,

hydrogen or organosilicon or substituted or unsubstituted carboorganic groups having from 1 to 30 carbon atoms, where two geminal or vicinal radicals R^1 to R^3 may also be joined to form a five- or six-membered ring, and R^1 , R^2 and R^3 in part or in whole, and independently of one another, are a group which carries a substituent

18

✓ attached via a carbon atom, in the α position relative to the nitrogen atom of the 1,3,5-triazacyclohexane ring, and

b) the alkylalumoxane.

13. A process as claimed in claim 12, wherein the olefin is ethene.

* * * * *